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Electrical characterization of molecular beam epitaxial GaAs with peak electron mobilities up to $\approx 4\times10^5$ $cm^2\ V^{-1}\ s^{-1}$

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The effect of varying the temperature (T_{cr}) of an $As_4 \rightarrow As_2$ cracker furnace between 600 and 700 °C on the properties of GaAs grown by molecular beam epitaxy has been evaluated using 4–300 K Hall measurements and 4.2 K far-infrared photoconduction spectroscopy, in an extension of earlier work on high-mobility material (Ref. 1). The residual donors are silicon and sulphur with mid-10^{13} cm^{-3} concentrations under As_2-growth conditions ($T_{cr} = 700$ °C). By lowering T_{cr}, the silicon concentration is reduced substantially, leaving sulphur as the principal impurity. A 15-μm-thick layer grown with $T_{cr} = 650$ °C has measured free-electron densities of $\approx 2.8\times10^{13}$ cm^{-3} and peak mobilities $\approx 4\times10^5$ $cm^2\ V^{-1}\ s^{-1}$ at $\approx$28–42 K, the highest ever recorded in bulk GaAs.

We have described recently the growth of n-type GaAs by solid-source molecular beam epitaxy (MBE) using arsenic dimers (As_2),[1] and the measurement of exceptional peak electron mobilities μ_{peak} between 2.75 and 3.32×10^5 $cm^2\ V^{-1}\ s^{-1}$. These values were comparable to the highest recorded for MBE-grown GaAs,[2,3] and for the best samples approached the highest peak mobility hitherto claimed in GaAs, 3.35×10^5 $cm^2\ V^{-1}\ s^{-1}$ in a 12-μm-thick layer grown by metalorganic chemical vapor deposition.[4] Growth with As_2 produced a marked reduction in the concentration of incorporated carbon compared to the level in layers grown under similar conditions with As_4 generated from the same batch of arsenic, resulting in lower compensation ratios for comparable free-electron densities. A further point noted in Ref. 1 was the consistency of the Hall data at 77 K, with the free-electron densities n_{77} and mobilities μ_{77} grouped around $\approx 1\times10^{14}$ cm^{-3} and $\approx 2\times10^5$ $cm^2\ V^{-1}\ s^{-1}$, respectively, despite the wide range of growth conditions investigated. This was tentatively explained by the inadvertent incorporation of Si from the cracker section of the As source. The Si concentration should therefore decrease as the temperature of the cracker section (T_{cr}) is lowered, with the possibility of further enhancements to μ_{77} and μ_{peak}. However, there could be an attendent risk of increased carbon incorporation as the proportion of As_4 in the total As flux increases and the As_2 component declines. The experiments summarized in Ref. 1 have now been extended to investigate the effect of varying T_{cr}. Unintentionally doped ("undoped") layers 15–30 μm thick have been grown with T_{cr} set at different values between 600 and 700 °C. The layers have been characterized by variable temperature Hall effect measurements in the range 4–300 K, and by 4.2 K far-infrared photoconduction spectroscopy (FIRPC). The original speculation concerning the origin of the Si is confirmed and, using T_{cr} as an adjustable growth parameter, the realization of the highest mobility ever in bulk n-GaAs is now reported.

Undoped layers were grown under (2×4)–(100)As stable conditions on to 2-in.-diam. semi-insulating (100)GaAs wafers, mounted without indium bonding in a Varian Modular Gen II MBE system described previously.[1] The arsenic sublimator of the As_2 source was set to a temperature of $\approx$290 °C to maintain a constant flux of As_4 tetramers into the cracker section. The cracking efficiency (defined as the percentage of As_4 tetramers converted into As_2 dimers) was estimated to be $\approx$70% at 600 °C, rising to $\geqslant$95% at 700 °C.[5] The growth conditions employed are summarized in Table I, where the wafer temperature (T_s) prior to growth was monitored with an optical pyrometer, and the flux ratio was determined from the ion currents generated by the Ga and As ($As_2 + As_4$) fluxes impinging separately on the beam monitoring ion gauge.

FIRPC spectroscopy of the Zeeman split hydrogenic donor transitions provides an accurate and accessible method of identifying individual shallow donor impurities in n-GaAs.[6,7] The samples for FIRPC analysis here were held in the Faraday configuration in a 10 T superconducting magnet and illuminated with radiation in the 40–600 μm range from a conventional optically pumped far-infrared laser, guided into the sample chamber of the cryostat by standard lightpipes. Band-gap illumination was employed to neutralize ionized donors thus narrowing the linewidths and further increasing the resolution.[8,9] Figure 1(a) shows the FIRPC spectrum of No. B48 taken with the 302 μm line which excites the $1s \rightarrow 2p_{-1}$ transitions. The three peaks are due to S, Sn/Se, and Si donors,[8] with the concentrations, N in the order $N_{Si} \gg N_S \approx N_{Se}$. The FIRPC lines are narrower than obtained previously from high-mobility MBE-grown n-GaAs,[10] an indication of the improved purity and higher 77 K and peak mobilities of this layer (see Table II). The FIRPC spectrum in Fig. 1(b) was recorded under the same conditions of temperature, bias, and band-gap illumination from No. B54, a 15-μm-thick layer grown with $T_{cr} = 650$ °C. The ratio of the S and Si peaks has changed such that $N_S > N_{Si}$ ($\gg N_{Se}$). The signatures of all three donors are better resolved and the

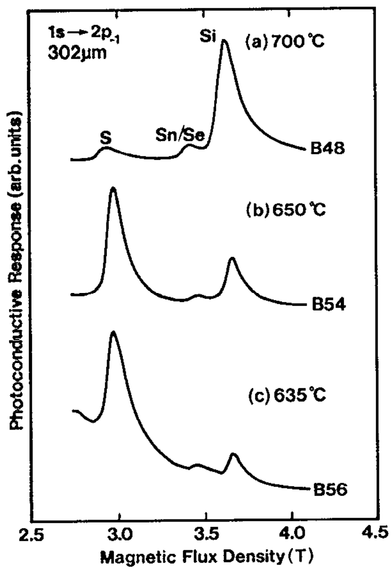

FIG. 1. Far-infrared photoconductive response vs magnetic flux density due to $1s \rightarrow 2p_{-1}$ donor transitions in three As_2-grown MBE GaAs samples under illumination with 302 μm radiation. The samples were grown with the cracker temperature set to (a) 700 °C (No. B48), (b) 650 °C (No. B54), and (c) 635 °C (No. B56).

linewidths for sample No. B54 are almost half those for No. B48, indicating an overall lowering in the concentration of each donor.

The trends in the FIRPC spectra are supported by variable temperature Hall data which was taken with a magnetic flux density of 0.2 T and with the applied electric field set low enough to avoid increasing the free charge densities by impact ionization. The free-carrier densities have been corrected for depletion effects at the surface and the substrate interfaces only.[11] Compared with No. B48, n_{77} for No. B54 has been reduced by a factor of 2.7, to 2.8×10^{13} cm^{-3}, while μ_{77} has increased to 2.2×10^5 cm^2 V^{-1} s^{-1}. The total donor and acceptor concentrations in this sample are estimated from theoretical data[12] to be $\approx 4 \times 10^{13}$ cm^{-3} and $\approx 1 \times 10^{13}$ cm^{-3}, respectively. A plot of mobility, μ as a function of temperature T in the range $\approx$4–300 K for No. B54 is shown in Fig. 2. The mobility peaks at the remarkably high value of $\approx 4.0 \times 10^5$ cm^2 V^{-1} s^{-1} over a broad plateau of temperatures from $\approx$28 to 42 K, the highest mobility ever recorded in GaAs irrespective of the growth technique employed, and exceeds 3.0×10^5 cm^2 V^{-1} s^{-1} from $\approx$9 to 65 K. Values of μ_{peak} in the range $3.8\text{–}4.1 \times 10^5$ cm^2 V^{-1} s^{-1} have been measured for several van der Pauw and Hall bar geometry

TABLE I. Summary of growth parameters.

Sample No.	Growth temperature (°C)	As:Ga flux ratio	Cracker temperature (°C)	Layer thickness (μm)
B48	611	3.2:1	700	15
B54	583	3.5:1	650	15
B55	583	4.1:1	600	20
B56	580	4.1:1	635	30

TABLE II. Summary of electrical properties of n-GaAs grown by solid source MBE with As_2.

Sample No.	n_{77} ($\times 10^{13}$ cm^{-3})	μ_{77} (cm^2 V^{-1} s^{-1})	N_a/N_d (Ref. 12)	μ_{peak} (cm^2 V^{-1} s^{-1})
B48	7.6	203 000	0.2	327 000 (42 K)
B54	2.8	220 000	0.2	402 000 (28–42 K)
B55	S.I.	Depleted	⋯	⋯
B56	S.I.	Depleted	⋯	⋯

samples prepared from wafer No. B54. Two other layers have been analyzed: Nos. B55 and B56 grown with $T_{cr} = 600$ °C and 635 °C, respectively. The $1s \rightarrow 2p_{-1}$ transitions for No. B56 [Fig. 1(c)], which are superimposed on the tail of the low field $1s \rightarrow 2p_{-1}$ peaks near 0.5 T, show a further reduction by a factor of 2 in the Si peak relative to S, in sympathy with the decrease of T_{cr} to 635 °C. No. B55 could not be measured because of contacting problems, and no Hall data could be recorded for either Nos. B55 or B56 which were believed to be fully depleted.

Although FIRPC spectroscopy does not allow a quantitative comparison of the donor concentrations from sample to sample, the peak ratios for a particular specimen are a reliable indication of the relative proportions of the individual donor species in that sample. If the density of donor-like intrinsic defects in these As_2-grown layers is low, then the contributions of Si(N_{Si}) and S(N_S) to the total extrinsic donor density (N_d^{total}) can be estimated by combining the FIRPC and Hall results, assuming also that the contribution from Se/Sn is negligible. The calculations, covering nine samples discussed both here and in Ref. 1, show that N_S is approximately constant at $\approx 3\text{–}5 \times 10^{13}$ cm^{-3} for $T_S \approx 580$ °C and an As_2:Ga flux ratio of $\approx$3.2:1. The concentration of sulphur incorporated into No. B48 is lower ($\approx 7 \times 10^{12}$ cm^{-3}) due to the higher value of T_S. The surface during growth was close to transforming from the (2×4)-(100)As stable to the (3×1)-(100)As-deficient reconstruction, when loss of S becomes significant via its reaction with free-surface Ga atoms and subsequent desorption as Ga_2S.[13] N_{Si}, in contrast, falls from $\approx 6 \times 10^{13}$ cm^{-3} in No. B48 (a typical value with $T_{cr} = 700$ °C) to $\approx 1.1 \times 10^{13}$ cm^{-3} in No. B54 ($T_{cr} = 650$ °C), and to an estimated 5.5×10^{12} cm^{-3} in No. B56 ($T_{cr} = 635$ °C). If the cracker section of the As_2 source behaves as a Knudsen-type source, then the Si flux will vary as $\sim p_{Si}(T)/T^{1/2}$, where $p_{Si}(T)$ Torr is the equilibrium vapor pressure of Si over Si at temperature T K, and from Ref. 14:

$$\log_{10} p_{Si}(T) = -20\,900/T - 0.565 \log_{10} T + 10.78.$$

N_{Si} is predicted to fall by $\times 13.8$ as T_{cr} is lowered from 700 to 650 °C, and a further $\times 2.3$ as T_{cr} is decreased from 650 to 635 °C. Both reductions are comparable to the estimates of $\approx \times 6$ and $\approx \times 2$ based on the FIRPC and Hall data, and provide confirmation that the Si flux does originate from a component in, or close to the cracker section of the

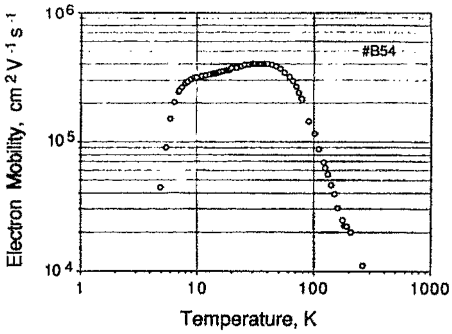

FIG. 2. Mobility μ vs temperature T for sample No. B54. The mobility peaks at $\approx 4\times10^5$ cm^2 V^{-1} s^{-1} between $\approx$28 and 42 K.

As_2 source. The total concentration of acceptors in the four samples examined here is estimated at $\approx 1\times10^{13}$ cm^{-3} or less, and no evidence has been found from 5 K photoluminescence measurements to suggest additional carbon incorporation as T_{cr} is lowered from 700 to 600 °C, and an As_4 component introduced into the As flux.[15]

In summary, n-GaAs with peak electron mobilities $\approx 4\times10^5$ cm^2 V^{-1} s^{-1} at $\approx$28–42 K has been grown by solid-source MBE using As_2. These are the highest values of μ_{peak} ever achieved in bulk n-GaAs regardless of the growth technique employed. Total impurity concentrations estimated at $\leqslant 5\times10^{13}$ cm^{-3} have been realized. FIRPC spectroscopy and variable temperature Hall effect analyses have demonstrated that Si from the As cracker contributes to the unintended n-type conductivity of the GaAs layers, although as T_{cr} is lowered below $\approx$650 °C, S incorporated from the solid As-charge becomes the major impurity. There may be scope for growing n-GaAs with even higher values of μ_{peak} using the 8N's Ga and 7N's As charges currently available. This would involve modifying the conditions used to grow No. B48, again setting T_s to 610–620 °C to initiate partial desorption of the incident S flux as Ga_2S, but at the same time operating the cracker at $T_{cr} \leqslant 650$ °C to minimize the Si contamination. A total impurity concentration of $\leqslant 1$–2×10^{13} cm^{-3} is a realistic target with the present MBE equipment.

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